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Graphite-tungsten twin limiters in studies of material mixing processes on high heat flux components

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Abstract

Graphite-tungsten twin limiters have been used at the TEXTOR tokamak for testing of high-Z metals as plasma facing materials and, in particular, for studies of the local and global transport of tungsten. The emphasis was on the change in surface morphology of limiters: the formation and properties of multicomponent co-deposits and the trapping characteristics of fuel on carbon and high-Z substrates exposed to the plasma under various operation conditions, i.e., heating scenarios, configuration of limiters, etc. Vast quantities of tungsten have been found to be locally transported to the adjacent graphite surfaces. Ion beam analysis also indicated strong intermixing of carbon, tungsten and boron on the hottest parts of the limiters. The results are discussed in terms of various mechanisms involving the transport of tungsten-containing species, possibilities of oxide production and formation of mixed (W–C–B) compounds. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Material mixing in controlled fusion devices can be described as the intentional or unintentional change in properties (chemical, fuel recycling, thermo-mechanical, etc.) of plasma facing components (PFC) resulting from the simultaneous use of several different elements in contact with the plasma. Mixing occurs due to the global and local transport of various species being eroded and re-deposited in the machine. It results in the formation of co-deposited layers retaining H isotopes [1,2]. Co-deposition has a serious impact on plasma density control, tritium inventory in D–T reactors [1–3] and production of dust particles [4]. Therefore, studies of material mixing on PFC are of considerable importance

when both low-Z and high-Z plasma facing materials (PFM) are to be applied in current and future fusion experiments [5].

For several years, tungsten has been tested as a divertor [6] and limiter [7] material in ASDEX-Upgrade and TEXTOR, which are operated with graphite as the main PFM. In the case of high-Z metals (e.g., Mo, W), local transport plays an important role in the migration of such species in the machine [8,9]. To study this effect in detail, the so-called twin limiter experiments have been initiated at TEXTOR [9,10]. The idea is to expose a limiter consisting of tiles made, in two equal parts, of graphite and tungsten. The basic goals to be achieved by applying those limiters are to recognize the impact of tungsten on the tokamak operation, to assess the erosion of various materials being exposed under the same conditions and to study the material mixing due to the erosion-deposition processes. The aim of this work was to determine the surface morphology of the twin limiter (in particular its graphite part), to assess the materials

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mixing due to the transport of tungsten and its impact on deuterium inventory, and the formation of mixed compounds.

2. Experimental

The experiments were performed with graphite– tungsten twin limiters of hemispherical (mushroom) shape and consisting of two equal parts: one made of graphite and the other made of tungsten, separated from one another by a 1.5 mm wide gap. This is shown in the schematic cross-section in Fig. 1(a). The limiters were introduced to the torus from the top of the vacuum vessel. A detailed description of the experimental set-up is given in [10]. The tungsten part was preheated to a temperature of about 450–500°C, i.e., above the ductileto-brittle transition temperature (DBTT) in order to prevent the limiter material from cracking caused by thermal shocks.

Three experimental campaigns with twin limiters were performed at TEXTOR. The ones described in this work were the second (Limiter 2) and third (Limiter 3) in that series. They were exposed to the Ohmically and neutral beam heated shots for 533 and 223 s, respectively. For exposure to the plasma, the top of the twin limiter was placed either at the minor ra-



Fig. 1. Twin limiter: (a) a schematic view in the toroidal direction; (b) gap area and indicated lines of ion beam analysis.

dius of 46 cm or less (45.0 cm), i.e., being immersed 1 cm into the plasma.

Surfaces of the exposed limiters were examined ex situ. Rutherford backscattering spectroscopy (RBS) was used for the determination of the tungsten, silicon and oxygen. Nuclear reaction analysis (NRA) was applied to quantify deuterium and boron originating from the boronization. The analyses were limited not only to the top part of the limiter but also the sides and the gap area were examined, as shown in Fig. 1(b).

3. Results and discussion

Exposures to the plasma resulted in distinct changes in the limiters' surface morphology due to the formation of co-deposits. The appearance of both halves differed distinctly. On tungsten, a thick blackish co-deposit was found only in the far end of the limiter and on the topmost part (3–5 mm wide) of the gap. The majority of the plasma facing surface, side surfaces and the rest of the gap area remained shiny. On the graphite part, in addition to the deposition zone at the far end, a shiny metallic layer was observed on the top, on the side surfaces and in the gap. The formation and composition of the deposition zones on tungsten limiters have previously been described [9]. The main emphasis of the studies reported in the following is on the surface state of the carbon part.

3.1. Top of the limiter

Fig. 2 shows the distribution and concentration of tungsten, boron and deuterium in the co-deposit formed on the top of the graphite part. These results, obtained when analyzing Limiter 2 along the central



Fig. 2. Distribution and concentration of tungsten, deuterium and boron on the plasma facing surface of the graphite part of the twin limiter.

line, are fairly representative, from the qualitative point of view, for both the limiters. Similar distribution patterns have been found for other scans made along the surface. The tungsten content reaches its maximum value near the tangency point. Afterwards, it steadily decreases and eventually comes down to a nearly constant value of about $0.8-1.3 \times 10^{16}$ cm⁻². In contrast, the concentrations of boron and deuterium are rather small close to the tangency point but they increase significantly, though not in the same manner, at larger distances. One noticeable difference between the deposition profiles of D and B is that the boron concentration close to the very top of the limiter is a few times bigger than that observed at a distance between 9 and 25 mm.

This complex deposition pattern is a net result of global and local transport phenomena. Erosion of tungsten occurs mainly via physical sputtering by plasma impurity ions: carbon, oxygen and boron. The majority of sputtered species are neutrals and, as measured with optical spectroscopy [10] and also modeled using a Monte Carlo code ERO-TEXTOR [11], the range of tungsten atoms is up to 12-15 mm (normal to the limiter surface) before they get ionized. Species ionized close to the limiter surface may promptly be re-deposited [8] within the first gyro orbit (Larmor radius is 1.4–2.0 mm under the experimental conditions). This explains local accumulation of large quantities of tungsten on graphite and on areas close to the tangency point. Deposited atoms are partly resputtered and transported again. The process leads to the 'spill over' of tungsten on graphite until the point of equilibrium between the re-deposition and re-erosion is reached. Global transport also contributes to the redeposition of tungsten. This involves species once removed from the limiter, ionized and transported in the torus. A fraction of that flux, approximately $3-10 \times$ 10^{13} cm⁻² s⁻¹, as measured with the collector probe technique in the scrape-off layer [12], returns to the surface. The presence of tungsten detected up to the far end and on the side surfaces of the limiter is attributed mostly to the deposition of such fluxes.

The co-deposited boron and deuterium (together with carbon impurity atoms, which could not be measured on graphite) originate only from the global transport. Their deposition profiles increase with the increased radial distance from the plasma. This is connected with high temperatures (over 2000 K) and resultant high erosion rates of D and B near the tangency point. The deposition starts to dominate in cooler zones. Ion beam analysis does not carry the information on the chemical state of the mixed layer, but the shape of the spectra indicate strong intermixing of carbon and tungsten, suggesting the coexistence of elemental C and W together with carbides [13] or mixed W–C–B compounds.

3.2. Gap area

Deposition profiles of tungsten detected in the gap area on the graphite part are shown in Fig. 3. The analyses were done along the lines shown in Fig. 1(b). Two distinct deposition regions are seen: a region with a very steep deposition profile close to the limiter top and another one with a constant tungsten concentration. In the topmost part of the gap, the tungsten content reaches a value of 8×10^{17} cm⁻² which is even a few times higher than that observed on the plasma facing surface of carbon (see Fig. 2). Within a distance of 5 mm, the concentration sharply decreases to the level of 1×10^{16} cm⁻² and then it remains constant. From the shape of the profiles, one infers that at least two different processes are responsible for the deposition of tungsten in that region.

The geometry of the limiter and the magnetic field lines allow us to exclude the possibility that tungsten ions transported in the torus could enter the gap and become deposited. The transport of neutral species is to be considered. In the narrow upper region, the deposition of tungsten may be attributed to the local transport of neutrals produced by sputtering at the very top of the limiter. The angular distribution of sputtered species follows a cosine dependence and, therefore, only a small fraction of neutrals may enter the gap and be deposited in a belt a few millimeters wide. The cosine distribution and the angled edges of the limiter top implicate very steep deposition profiles. This is exactly observed in the experiment. Atoms once deposited in the gap and 'hidden' from the direct impact of plasma fluxes are not re-sputtered, and a build-up of the layer may proceed step-by-step. This would explain why the resultant net deposition in the upper part of the gap is significantly higher than that found on the plasma facing surface, where the re-deposited tungsten undergoes further erosion, as discussed in Section 3.1.



Fig. 3. Distribution and concentration of tungsten in the gap area on the graphite part of the twin limiter. Lines of analysis are shown on a schematic view of the gap area.

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To produce a fairly flat and uniform deposition profile deep in the gap, one needs an uniform spatial distribution of tungsten species. The transport of sputtered neutrals due to a multiple reflection in the gap is considered to be negligible or highly improbable, because the energy of sputtered tungsten neutrals is low, with the maximum in the energy distribution at around 4.5 eV. This effect is considered to be insignificant, because the measurements performed for several metalcarbon systems have shown very high sticking probability (0.98-1) of sputtered atoms to graphite [14]. Multiple reflection would then occur only for high energy (above 100 eV) sputtered neutrals, but the fraction of such species does not exceed 1%. Even assuming the scattering of tungsten neutrals on the already existing tungsten film, one would expect the non-uniform (e.g., exponential) deposition profile limited to a few millimeters, but not centimeters as observed in the experiment.

Energetic neutrals are also produced in chargeexchange collisions. These species may enter areas inaccessible to plasma ions. The flux of such neutrals reaching the gap be would composed of both deuterium and minority atoms present in the plasma, like boron, silicon, Inconel components (Ni + Cr + Fe + Mo) and tungsten. However, only trace quantities of Inconel elements are detected with ion beam analysis, while the concentration of tungsten is higher than that of deuterium or boron. This result indicates that another mechanism producing tungsten in the gaseous form might be decisive: sublimation of tungsten, discharge in the gap due to local electric fields or formation of volatile oxides. As the heat of vaporization (775 kJ mol⁻¹) is high, the sublimation of significant W quantities would require temperatures exceeding the melting point (3683 K), but no surface melting was observed on tungsten in the gap region.

The second hypothesis involves an electric discharge, which would locally generate a plasma of high carbon and tungsten content. This would explain the uniform distribution of tungsten on the graphite surface. The process might be of importance especially when the limiter was immersed by 1 cm into the plasma. Unfortunately, it is difficult to prove this hypothesis experimentally because no direct spectroscopy observations of species in the gap could be performed. Also the concentration of carbon on the tungsten part, in the gap area, could not be measured with ion beam techniques, because the sample was too heavy to be mounted on a vertical manipulator in the surface analysis station. From the colorimetric analysis of the carbon deposit on tungsten, one infers approximately 5×10^{17} C at cm⁻² (corresponding to 80 nm thick layer) in the topmost part of the gap, but no deposit is observed on other areas. However, C concentrations below 7×10^{16} cm⁻² cannot be detected with that method.

The ion flux to the limiters at TEXTOR contains oxygen at the level of 1.0-1.5% of the total flux, i.e., $7-10 \times 10^{16} \text{ cm}^{-2} \text{ s}^{-1}$, a few times lower fluxes are observed only after the fresh boronization or siliconization [15]. Tungsten shows high affinity to oxygen and it forms a number of solid and volatile oxides stable at high temperatures [16–19], but the formation of tungsten (VI) oxide, WO₃, and tungsten (IV) oxide, WO₂, is very unlikely to take place because the oxygen content is too low in the strongly reducing environment. However, as there is a source of oxygen in the system, the formation, diffusion and reduction of oxide species may play a certain role in the transport of tungsten. The formation and diffusion of oxides would also partly explain the W deposition on the side surfaces of the limiter. The above considerations lead to a conclusion that the local electric discharges and, to a small extent, the formation of oxide species are key steps in the tungsten erosion and re-deposition in the gap.

4. Summary and conclusions

Material mixing occurring on surfaces of graphitetungsten twin limiters has been studied. It has been shown that eroded tungsten, to a large extent, migrates locally to the adjacent plasma facing surfaces. The major mechanism of migration is related to the prompt redeposition of sputtered species. On the hottest parts of the limiters, the formation of mixed W–C–B compounds could be inferred from the ion beam analysis spectra and deposition profiles of those elements. Deposition of tungsten in gaps, i.e., areas hidden from the direct impact of plasma fluxes, has been observed. One concludes that the erosion caused by local electric discharges is the most probable mechanism for the metal migration and deposition on surfaces located in gaps between the limiter tiles.

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References

- [1] G. Federici et al., J. Nucl. Mater. 266-269 (1999) 14.
- [2] M. Rubel et al., J. Nucl. Mater. 266-269 (1999) 1185.
- [3] C.H. Skinner et al., J. Nucl. Mater. 241-243 (1997) 887.
- [4] J. Winter, Plasma Phys. Control. Fusion 40 (1998) 201.

- [5] R. Parker et al., J. Nucl. Mater. 241-243 (1997) 1.
- [6] K. Krieger, R. Neu, The ASDEX Upgrade Team, J. Nucl. Mater. 266–269 (1999) 207.
- [7] J. Rapp et al., Plasma Phys. Control. Fusion 37 (1997) 1691.
- [8] D. Naujoks, R. Behrisch, J. Nucl. Mater. 220–222 (1995) 227.
- [9] M. Rubel, V. Philipps, A. Huber, T. Tanabe, Phys. Scripta T81 (1999) 61.
- [10] T. Tanabe et al., Fus. Eng. Des. (in press).
- [11] Kirschner, V. Philipps, J. Winter, U. Kögler, Nucl. Fus. 40 (2000) 989.
- [12] M. Rubel et al., J. Nucl. Mater. 249 (1997) 116.

- [13] Gmelin Handbook of Inorganic and Organometallic Chemistry, Tungsten Suppl. vol. A 5b, 8th Ed., Springer, Berlin, 1993 (Chapters 7–9).
- [14] Emmoth, H. Bergsåker, Nucl. Instrum. and Meth. B 33 (1988) 435.
- [15] U. Samm et al., J. Nucl. Mater. 220-222 (1995) 25.
- [16] Gmelin Handbuch der Anorganischen Chemie, Wolfram, vol. B 2, 8th Ed., Springer, Berlin, 1979, pp. 1–31 (Chapter 3.2).
- [17] L. Brewer, Chem. Phys. 52 (1953) 2.
- [18] P.E. Blackburn, M. Hoch, H.L. Johnston, J. Phys. Chem. 62 (1958) 769.
- [19] L. Brewer, G.M. Rosenblatt, Chem. Rev. 61 (1961) 257.